Uncertainty: A Discriminator for Above and Below Boiling Respository Design Decisions

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The US nuclear waste disposal program is evaluating the Yucca Mountain (YM) site for possible disposal of nuclear waste. Radioactive decay of the waste, particularly spent fuel, generates sufficient heat to significantly raise repository temperatures. Environmental conditions in the repository system evolve in response to this heat. The amount of temperature increase, and thus environmental changes, depends on repository design and operations. Because the evolving environment cannot be directly measured until after waste is emplaced, licensing decisions must be based upon model and analytical projections of the environmental conditions. These analyses have inherent uncertainties.

There is concern that elevated temperatures increase uncertainty, because most chemical reaction rates increase with temperature and boiling introduces additional complexity of vapor phase reactions and transport. This concern was expressed by the NWTRB, particularly for above boiling temperatures. They state that "…the cooler the repository, the lower the uncertainty about heat-driven water migration and the better the performance of waste package materials. Above this temperature, technical uncertainties tend to be significantly higher than those associated with below-boiling conditions." (Cohon 1999).

However, not all uncertainties are reduced by lower temperatures, indeed some may even be increased. This paper addresses impacts of temperatures on uncertainties.

Uncertainty Analyses for Thermal-Hydrological-Chemical (THC) Processes

Significant sources of uncertainty in THC analyses include incomplete data on rate constants and uncertainties in effective reactive surface areas. Elevated temperatures increase reaction rates following the Arrhenius activation energy principle (e.g., Wolery, 1999). Thus, the divergence from and the length of time to reach equilibrium would be minimized at higher temperatures. If a reaction approaches equilibrium rapidly, relatively large errors in rate law parameters, and possibly even reactive surface areas, may become insignificant. Such a reaction might properly be modeled as approaching equilibrium instantaneously, thus, obviating the need for kinetic data. The equilibrium state depends only on thermodynamic data, which are known better than kinetic data. Therefore, there is less uncertainty in assessing equilibrium chemical processes than kinetic processes.

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To the extent that relevant thermodynamic data are better known than kinetic data, analyses that require kinetic data have increased uncertainty. However, it does not follow that kinetic data are less well known for the higher temperatures. Much of the data on kinetic rates has been obtained in experiments at higher, rather than lower, temperatures (to shorten the time for reactions to be observed and to boost signals above detection limits). Thus, there are more directly measured kinetics data for the $70\text{-}100^0C$ range of temperatures, and therefore less uncertainties, than for temperatures between $30\text{-}70^0C$ where data is extrapolated.

Finally, increased aqueous process reaction rates with higher temperatures only apply when liquid water is present. At near or above boiling point temperatures water is removed and aqueous reactions cease. Reaction rates increase rapidly up to the boiling point, where they drop to zero. When water is present at elevated temperatures, both reaction rates and associated uncertainties are high. When there is no water, the rates and associated uncertainties drop to zero.

While uncertainties regarding aqueous reactions may decrease for the "above boiling case", there remains additional complexity of vapor dominated geochemical reactions. Before a decision regarding the magnitude of relative uncertainty can be made, the impacts of vapor interactions must be considered. Because the mass of water in the vapor phase is limited, alteration is not expected to be significant.

Uncertainty Analyses for the Thermal-Hydrological (TH) Processes

For above boiling conditions there are uncertainties about the redistribution of water that will be vaporized. Water within the matrix is relatively immobile. However, when the water is vaporized it can travel readily within the matrix/fracture system until it either escapes or condenses in cooler regions of rock. Questions arise as to the fate of this condensed water. The NWTRB note that, "…the understanding of water mobilization and migration processes and effects … is still far too limited to engender a reasonable degree of confidence…" (Cohen, 1999).

Flow in unsaturated media involves two phases, liquid water and air/water vapor, which adds uncertainties beyond those of saturated flow. YM analyses must consider two-phase flow, regardless of thermal conditions. Heat vaporizes water, which creates a more vapor dominated two-phase flow condition. Boiling may introduce uncertainties in the extent that vapor phase dominates and therefore changes the relative conductivities for each phase. However, higher thermal loading scenarios may reduce uncertainties because the vapor phase becomes so dominate. Volume increases associated with phase change of water from liquid to vapor will cause water vapor to occupy most, if not all, of the fracture and matrix porosity, minimizing two-phase flow conditions.

Vapor dominated systems may reduce other TH process uncertainties. Viability Assessment peer review (CRWMS M&O 1999, p. 47) noted that use of van Genuchten "…model is not justifiable…", because it ignores a number of processes, including: unstable nature of gravity-driven infiltration; hysteresis during episodic flow, small scale

(sub-grid) heterogeneities; effects of connectivity of fracture and matrix continua; and effect of abrupt changes in properties on transport fluxes. Further, that the fracture-matrix factor, while reasonable and justified to account for a variety of processes for which the physics are not directly incorporated in the models, is "…devoid of convincing physical meaning… and reflects a lack of understanding of the …physics of the process … The AFM [Active Fracture Model] is intuitive, but is based essentially on inverse modeling to match saturation data from the field and not on a physical basis from laboratory studies, etc…" (CRWMS M&O 2000a, Section 3.4.1.4.1). During active boiling periods, when vapor occupies most or all porosity, many of these factors, and associated uncertainty, including fracture-matrix interactions, active fracture model approach, gravity driven infiltration, and even heterogeneity and fracture connectivity issues are either eliminated or greatly reduced.

Another TH uncertainty is the fate of thermally mobilized water. Does it drain out of the system or increase saturations, particularly above repository drifts, so that water can then flow through fractures and contact the waste? Prior to boiling, most water is in matrix pores where it would be relatively immobile. After vaporized it would condense mostly in fractures, where it would be relatively mobile. Of particular concern is whether the condensate can flow through the fractures with sufficient volume and velocity to penetrate the above boiling zone and contact the waste packages before the water is vaporized. Field and laboratory tests indicate that this is possible under some conditions.

For below boiling temperatures, significant uncertainty in drift seepage estimates results from the natural hydrologic system heterogeneity. Seepage likely occurs in only a small percentage of the drift area and from only a small percentage of the fractures. It is highly dependent on hydrologic properties, which can vary by orders of magnitude. Thermal properties are much better constrained. Thus, seepage into above boiling drifts is much more predicable than it is for below boiling drifts. Furthermore, the volume of dried rock surrounding the drifts would tend to imbibe any water from percolation events so that seepage would be minimized. After a few hundreds to thousands of years, when temperatures decay, the above boiling repository would have the similar seepage uncertainties as an originally below boiling repository.

Another area of uncertainty relates to the potential for heat-pipe formation. Green et al. (1991, pp. 164 to 165) reporting on laboratory experiments state " …. A heat-pipe process will occur only in the matrix block nearest the heat-generating source during the early stages after waste emplacement. Liquid water will only cross the fracture [to the next matrix block] when the moisture … of the matrix …is sufficiently high… to cause the fracture to saturate. Prior to this, the matrix block on the heated side of the fracture will desaturate thus denying the heat-pipe process the return flow of liquid water necessary to continue." Manteufel et al. (1993, p. 577) conclude that heat pipes will be negligible because of the low matrix permeability. Field test results address these issues, but the issue is fracture- matrix-block-geometry specific. Below boiling temperatures reduce or eliminate heat-pipe-related uncertainties.

<u>Uncertainties Associated with Interactions with Engineered Components of the System</u>

Environmental conditions impact uncertainties in analyses of corrosion of waste packages and dissolution and mobilization of radionuclides. The most critical environmental factors to the performance of these components are the amount of water that can seep into the drifts and the chemistry of that water.

The chemistry of any water that might seep into drifts and contact waste or waste packages will evolve from rock-water interactions mainly involving water flowing in fractures. For below boiling designs, both pore water and percolating water would remain to interact with NF rock at elevated temperatures because of kinetic processes. The composition of percolating water has not been measured, and therefore, evaluations of water chemistry for the below boiling case must consider kinetic processes with water whose chemistry is not well understood. For above boiling designs, the NF rock would dry rapidly and rock-water interactions in that region would be minimal to non-existent. Rock-water interactions would mostly occur some distance from the drifts. In these zones water that interacts with the rock will be mostly dilute condensate deposited in the fractures. If the condensate is imbibed into the matrix, it will tend to dilute the pore water but the ultimate water chemistry will likely be determined by the rock-water interactions over long times therefore approaching equilibrium. The chemistry of water in the fractures will be dominated by rock-water interactions with very dilute condensate. The less well characterized percolating waters will not become dominant over condensate until the temperatures drop significantly and reaction rates will be lower. Thus, uncertainties in water chemistry will be lower for the above boiling case, at least during the period of boiling, and possibly beyond that time frame.

Other important environmental interactions that contribute to uncertainty involve in-drift temperature and relative humidity. The presence of liquid water (or high humidity) when temperatures are elevated constitutes the most aggressive conditions for container corrosion. For a below-boiling design, the potential for high humidity and potential liquid water contact at a time when temperatures are elevated is greater than for an above boiling case. The reason is that when drift temperatures (or more specifically thermal gradients) are high, the relative humidity is low, and any water dripping into the drifts will be evaporated or boiled away. Numerical models, as well as field and laboratory studies, indicate that at YM water will not return to the dried out region (with exception of higher percolation flux which could penetrate the thermal regime) until heat decays, and furthermore, that for many cases temperatures decay more rapidly than water returns (CRWMS M&O 2000b). As a result, temperatures may be lower for an above boiling case by the time moisture returns than for a below boiling case would where moisture will be present in the drift and near-field rock for the entire thermal heating period.

<u>Conclusions</u>

Some uncertainties associated with THC may not be reduced (may even be elevated) by below boiling designs. The system will be further from equilibrium for a below boiling design. Kinetics data may be better constrained near boiling than at intermediate

temperatures.   Furthermore, many of the THC uncertainties remain with lower repository temperatures; it is just a question of where in the system the processes occur. For above boiling drift wall temperatures, water is driven away and the aqueous THC processes are minimized or no longer active. Beyond the above boiling region the same THC processes would occur as for a below boiling repository, they just occur further into the rock mass, or at later times. If there is an extensive boiling region, there will be the addition of condensate, with different water chemistry, but there is no fundamental difference in the model and analysis of the THC processes.

Some TH processes, such as seepage during the early times of heating, will be less likely in an above boiling case. As such the uncertainties associated with this factor and with the associated heterogeneity will be less for an above boiling case. However, there are additional uncertainties introduced by the mobilization of water.

This introduces another factor to be considered when making the trade-off decisions. That factor is the consequence of errors in the assessments. For the below boiling case, if there are errors in the assessments of coupled processes, the errors occur within the region of the waste. For above boiling case, any errors in the coupled process assessment would apply to a region removed from the drifts. Therefore, it is possible that the rock mass between the zone of active coupling and the drifts would serve to moderate the consequences.

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